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The Study of the Hydrodynamics of Single Bubble Sonoluminescence

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for

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Abstract

The study of sonoluminescence has been under taken to determine the mechanisms for the production of the short burst of light that arises in an acoustically driven water cell. The investigations have reached a state of understanding where the underlying physical processes causing the conversion of acoustic energy to radiation can now be successfully simulated. Further, the effort has led to substantial outreach to the community including undergraduate student, post-graduate students, and professors. Finally, the experimental program has provided important information on the region where sonoluminescence works.

Introduction:

Single Bubble Sonoluminescence (SBSL) is the production of visible light by a gas bubble that is suspended in a fluid (normally water) by an acoustic standing-wave field. Present understanding of the phenomenon suggests that SBSL may result in temperatures of over 10^5 K (which approaches the temperature found in the solar corona), pressures of over 10^{12} Pascals (close to the pressure at the center of the planet Jupiter), light emission of less than 10^9 s duration, and concentration of mechanical energy of up to 10^{12}. In SBSL, a 10-μm-diam bubble (i.e., a bubble with a diameter of about 1/10 the width of a human hair) oscillating in an audiofrequency (25 kHz), ultrasonic field synchronously emits on the order of a million photons in a short pulse each acoustic period. The process by which the acoustic energy is converted into electromagnetic energy is not completely understood and is being investigated by numerous research groups around the world.

In this project, we are addressing several important topics of general interest to LLNL, including a detailed study of the hydrodynamics in spherical geometry, the properties of matter at extreme conditions, and the source of the light flash. Our methods include: (1) experiments that look at the hydrodynamic collapse of a bubble; (2) simulations of the spherical nature of the collapse—the details of the simulations that must be exercised can be rigorous tests of the hydrodynamic codes; (3) studies that have impact on the equation of state of the gases, in the bubble as well as that of the water at extreme conditions—necessary because there was no reasonable solution of the source of the emissivity from the bubble; (4) unique techniques such as ultrafast-laser probing of the collapsed bubble—which will provide basic information on the ability of codes to map out the implosion phase correctly; and (5) studies aimed at understanding the light emission—these may provide a path for the use of this rather interesting phenomenon.

We have successfully engaged high-quality students in our project. These young bright individuals are attracted to LLNL to participate in research in which they become fully involved. This project has been an exceedingly effective method for introducing researchers to LLNL and for gaining public awareness.

Our experiments and simulations during FY 1999 essentially confirm that the light flash is caused by plasma formation. To reach this conclusion, we studied the behavior of the light emission from a well-characterized cell. We (1) set up a laser-probing system to obtain images of the bubble with 10-ns time resolution—this is used to measure the maximum and ambient bubble radii during the acoustic cycle, (2) assembled a thermoelectric system to allow control of the water temperature—this temperature is critical to the amount of light emission from the bubble, (3) used an optical streak camera to measure the duration and time shape of the light emission from the bubble, and (4) investigated the pulse duration as a function of water temperature, driving pressure, and gas concentration—we observed 50- to 300-ps duration pulses. Our measurements provided data that rigorously tested the hydrodynamic models.

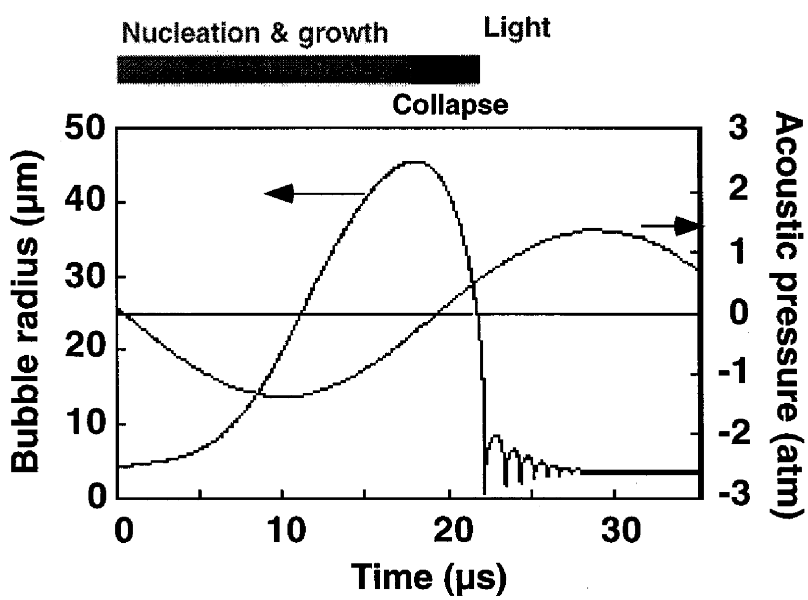

Figure 1: The time history of the sonoluminescence process showing the bubble radius on the left and acoustic drive pressure on the right abscissa versus time. The overall sequence is schematically labeled above the plot.

Brief Description of Single Bubble Sonoluminescence

To set the stage for the our most recent development a very brief outline of the sonoluminescence process is presented here. For the work performed here, the broad overview is that a bubble is created in a glass flask of water that collapses to very small diameter and emits a burst of light. In slightly more detail the bubble is formed by the application of a hot wire in the flask near the center. There are acoustic drivers on the flask that operates at a frequency of approximately 33 kHz. This creates a pressure wave taking the bubble that has a maximum radius, $R_{max} \sim 50$ μm, to a converged radius, $R_{min} \sim 0.5$ μm. The energy concentration in this process is quite large, on the order of 10^{12}. At the end of the converging phase of the bubble there is a light burst of 50 to 300 ps in duration, which depends sensitively on the water temperature, gas concentration in the water, and drive pressure. The process repeats each acoustic cycle. The entire history of the process is shown in figure 1.

Results of the Investigations

In the following we report on the three major aspects of the project. First we will cover the outreach that this project has generated. Here we will present the contacts that sonoluminescence has allowed us to make with the wider community. Second, we will discuss the theoretical program where we have managed to explain the nature of sonoluminescence for the first time, explaining all of the trends that have been observed. Finally, we discuss the experimental program where we have mapped out the dependencies of sonoluminescence on acoustic drive pressure, temperature, and gas concentration.

Technical Outreach

In this area we have made contributions in several areas that are critically important to the scientific well being of the laboratory. These are:

Undergraduate Education

We have managed to attract four students for training on the project. The nature of the sonoluminescence experiment is simple so that it allows undergraduates to make a contribution while learning the techniques necessary to perform experimental research. The students, D. Froula, M. Schnittker, D. Solis, and B, Whitney all participated in the experiments and provided meaningfully to the project with extended periods (> 6 months) in most cases spent at LLNL.

Collaborations with Universities

In the area of university collaboration we have formed strong ties with the at least five institutions. First, and foremost we have formed a partnership with the experimental group of Professor T. Matula at the University of Washington Applied Physics Laboratory. Professor Matula has received Presidential Young Fellow and a DOE Young Scientist Awards for his work on sonoluminescence.

Further, we are actively work with groups at Yale to simulate the high quality data from noble gas bubbles that has been obtained there. These data are of interest as all parameters have been carefully kept constant from one noble gas experiment to the next so it can provide a rigorous test.

Next, we have worked with researchers at University of California at Berkeley and the National Center for Physical Acoustics at the University of Mississippi on simulations. Finally, we have forged a collaboration with the Oxford University to model spectral emission from the bubble. This work has blossomed and we have had both Post

-Doctoral visitors and short term student visitors working on the experimental and theoretical aspects of the emission.

Technical Papers, Technical Presentations and Popular Press Articles

We have worked to disseminate the information that has been developed during the project. There have been three major publications, and four presentations at scientific meeting. The papers are:

- Froula, D. and P. Young, *Sonoluminescence for the Undergraduate Laboratory*, UCRL-JC-129336. 98-ERD-081.
- Matula, T., I. Hallaj, R. Cleveland, L. Crum, W. Moss, R. Roy, R., "The Acoustic Emissions from Single-Bubble Sonoluminescence," *J. Acoustic Soc. of America*, **103**, 1377, UCRL-JC-131348. 98-ERD-081.
- Hilgenfeldt, S., D. Lohse, W. Moss, "Water-Temperature—Dependence of Single Bubble Sonoluminescence," *Phys. Rev. Lett.*, **80**, 1332, UCRL-JC-131347. 98-ERD-081.
- Moss, W. C., et al., "Computed Optical Emission from a Sonoluminescing Bubble," *Phys. Rev. E*, **59**, 2986, UCRL-JC-131765. 98-ERD-081.
- Moss, W. C., J. L. Levatin, A.Szeri, "All good oscillations must come to an end," *Science*, UCRL-JC-134371. 98-ERD-081.

Further, the meetings to which we have presented work are:

- 137th Meeting of the ASA and 2nd Convention of the European Acoustics Association, Berlin (3/99) "Computed Optical Emissions from a Star in a Jar" UCRL-JC-131765
- DARPA Workshop on Sonoluminescence, Washington DC, (11/98) "Star in a Jar" UCRL-MI-128221

- 51st Annual Gaseous Electronics Conference and 4th International Conference on Reactive Plasmas, Maui HI(10/98) Modeling the Plasma in a Sonoluminescing Bubble" UCRL-JC-131009
- International Congress on Acoustics and Acoustical Society of America Meeting, Seattle, WA (6/98) "Computed Spectral and Temporal Emissions from a Sonoluminescing Bubble" UCRL-JC-128850

One of the important aspects of sonoluminescence is the fact that it has over the past decade attracted much interest by the lay public. In this way we have made two contributions to the popular press.

- First, we have been broadly quoted in a cover story for Popular Science for December 1998.
- Second, we have been quoted on the MSNBC world wide web site as an interview and article on 3/31/99.

Advances in Simulation Capability

The state of the art calculations in predicting the trends for sonoluminescence are now well advanced by the work paid for in part by this LDRD project. In brief we have shown by extensive simulations and comparison with extant data that the SBSL arises from a cool dense plasma. This is a major achievement that essentially ends the controversy that has arisen over the decade to explain the conversion of the acoustic drive to light emission. Further, we have advanced the predictive modeling of the SBSL for the case of a xenon bubble in water. Indeed, certain aspects of the Xe emission can be experimentally observed. Finally, we have developed with the Oxford group a detailed plasma spectroscopic model for conditions relevant to SBSL, that is a state of warm dense matter. This has led to the possibility that one could soon improve the current modeling currently carried out in the radiation hydrodynamic simulations.

Trends now Well Modeled

The calculations have been rigorously compared to available data we now find that the trends in the data are in agreement with our simulations. To achieve this level of agreement we have improved the emission and transport models and incorporated them into the radiation hydrodynamics simulations performed using Lasnex. The constituents of the bubble are now described by a mixed water vapor and noble gas EOS which was created especially for this regime

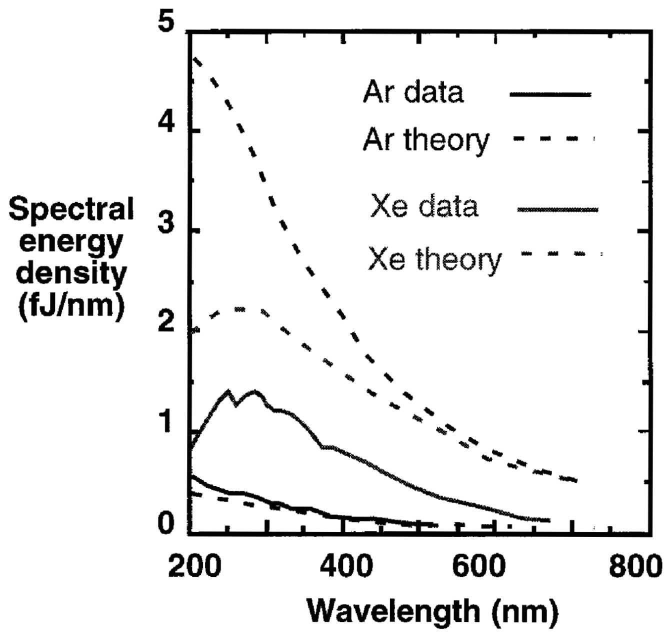

Figure 2. The comparison of argon and xenon filed bubbles showing the ability of the simulations to predict the broad spectral features correctly.

Our model shows the same trends as experimental data in three critical areas of comparison. First, the shape of the emission from the bubble as a function of time is asymmetric as measured. The simulated emission shows a fast sharp rise followed by a slower fall and ends with a low amplitude tail. Second, the temporal width of the pulse is now predicted to depend on liquid temperature and driving pressure as measured. Indeed, the simulations show that the duration of the pulse for a xenon filled bubble is greater than that for an argon filled bubble which in turn is predicted to be greater than that for a bubble filled with an air mixture. These relative durations have been observed. Finally, the spectrum of the emission is shown to peak at 300nm for a xenon bubble and at less than 200 nm for an air (or argon) bubble as observed.

In the figure 2 we show an example of the quality of the simulations. Here we see that we show the cases for both argon and xenon filled bubbles. Since the emission data shown do not have simultaneous bubble dynamics measurements thus we have to assume an ambient bubble radius, R_0. In the case of the argon filled bubble we show tow cases one for a $R_o = 2$ μm , and one for 6 μm. For the xenon case we estimate the bubble R_0 of 4 μm. In both cases the overall features are well reproduced and we have explained the xenon peak for the first time.

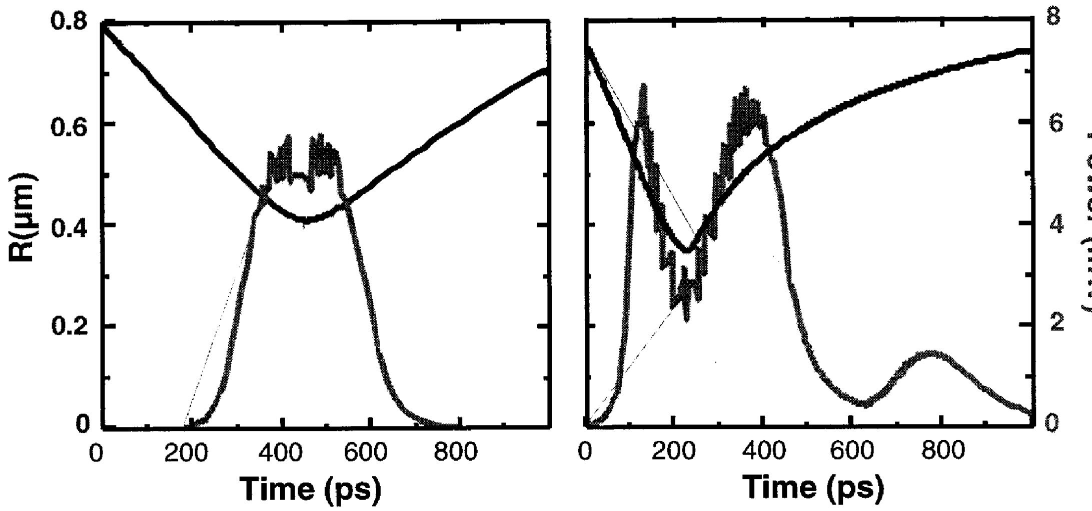

Figure 3. The time history of the radius, in blue, on the left hand abscissa and the power emitted, in red, on the right hand abscissa for a xenon bubble driven at two different drive pressures. The graph on the left represents a xenon bubble driven with a low pressure while the one on the right is driven at a high pressure. The distinct emission signals could be a critical test of the modeling capability.

Prediction can now be made with Model

We now have enough confidence to begin to use our model to make predictions. As an example we show below in figures 3 simulations for high drive pressure and low drive pressures on a bubble filled with xenon. As we can see from a comparison of the two figures 3 the optical pulse intensity as a function of time is a sensitive measure of the drive pressure. Note that even though we have calculated the pulse widths and spectra of the xenon SBSL emission. The sensitivity is due to the fact that the bubble which agree with data the dynamics equation of state and the opacity all effect the emission sensitively so that different drives yields different time histories. These prediction could be tested in the laboratory providing further information on the simulation capability and the underlying physical models that are necessary.

Spectroscopic Capability Developed

The background of the model is based on work performed when we were asked to look at a paper by LePointe "Nature of the 'extreme conditions' in single sonoluminescing bubbles" Journal of Physical Chemistry, **100**,12138 (1996) The paper attempted to explain the SBSL emission but did not use a consistent set of arguments. T make a spectral synthesis model we require three things: 1) the atomic structure data and any rates necessary; 2) the method for determining the level populations; and, 3) a prescription for turning these data into a synthetic spectrum. This third element includes formulations of the bound-bound line shapes, bound-free edges, and an ionization potential lowering model – to mimic the reduction of the principal quantum states available in the plasma etc.

The atomic structure data for low Z elements can be found in the TOPBASE data base. The argon neutral and single ionized atomic data is available and with a suite of codes we can take the extracted data and put it in a usable form.

Second, due to the rather high density we assume that the level populations are in local thermodynamic equilibrium (LTE) which entails invoking the Boltzmann equation for the ratio of the level populations in a particular ion stage and the Saha-Boltzmann equation to relate the populations ratios of the ion stages.

The third element is achieved in final step by a straightforward generation of the spectrum including bound-bound, bound-free, and free-free processes. Note that recombination processes are thereby included.

We can see a sample of the results of the spectral synthesis compared to the spectral radiance of the bubble emitted into 4π, i.e., W/nm measured by Hiller et al., in figure 4. Spectral line broadening plays an extremely important role at the assumed densities achieved by the gas in the bubble, it makes the myriad bound-bound transitions look like continua and making the recombination continua the dominant transitions in the sonoluminescence case. In figure 4 there is a comparison with data of calculations for Ar emission from a 2 μm radius sphere emitting for 50 ps at 33 kHz. These parameters are selected as a first estimate. Thus, we ignore gradients and temporal behavior in this comparison.. We show the spectra at several plasma conditions and also the measurements taken from Hiller et al. There are several things to note.

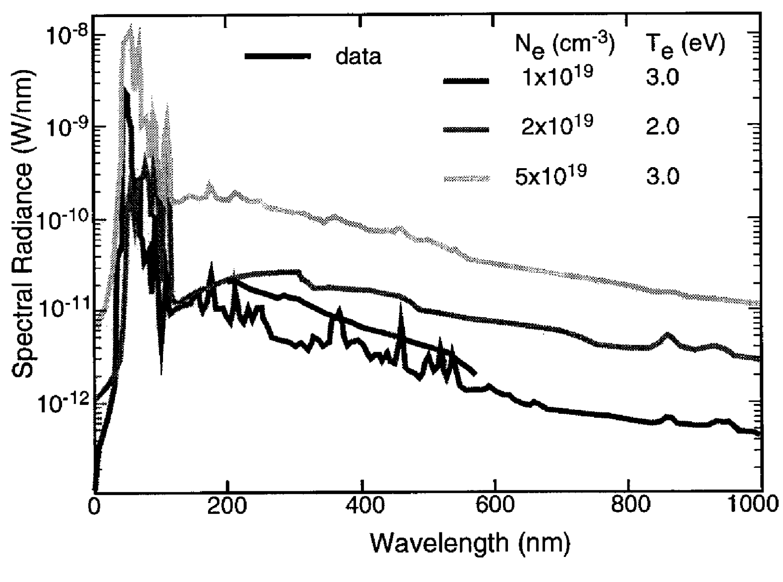

Figure 4. The spectral emission from SBSL driven at 33 kHz and emitting for 50 ps from a uniform 2 μm radius sphere. The black line indicated the measured spectrum while the three calculated spectra (blue, red, and green lines) are for different conditions to

illustrate the variation of the spectrum with temperature and density. The line below 200 nm will be masked from observation due to the large optical depth of the water.

First, the bound-free emission is a dominant part of the spectra and can be clearly seen in the spectral region between 200 and 300 nm. However, there are numerous other continua that are obscured by bound-bound transitions. Second, the figure indicates that to obtain a smooth spectrum one either has to have high density (see the spectrum for $5x10^{19}$ cm^{-3}) or low temperature (see any spectrum at 2 eV). Third, one can see that at low density (where the lines are somewhat narrower) and higher temperature the line transitions start to come up (see the spectrum for $5x10^{19}$ cm^{-3}, 3 eV). Here one observes a few narrow features; but, there are actually over 3600 transitions in the spectrum..

It is clear from this simple first simulation of the spectrum that more work could be profitably performed.

Experimental Results

Experimentally we have achieved the first phase of the work. This can be summarized by the following facts. First, we have performed measurements of the bubble emission characteristics with an emphasis on making the emission reproducible and reliable over a wide range. That is, we have measured the pulse duration, the pulse intensity and the bubble physical size, i.e., the bubble ambient radius (R_0) and the bubble maximum radius (R_{max}), over the physical parameter space. The measurements include variations of the drive pressure, temperature, and, gas concentration. This has led to the ability to map out the sonoluminescence space in greater detail than as been possible previous. In figure 5 we show the intensity of the SBSL emission versus the water temperature and the drive pressure. The intensity scale indicated by the color scale to the right hand side of the plot. Note that there are clear demarcations where the SBSL work and where is does not and these are indicated by the white line on the figure.

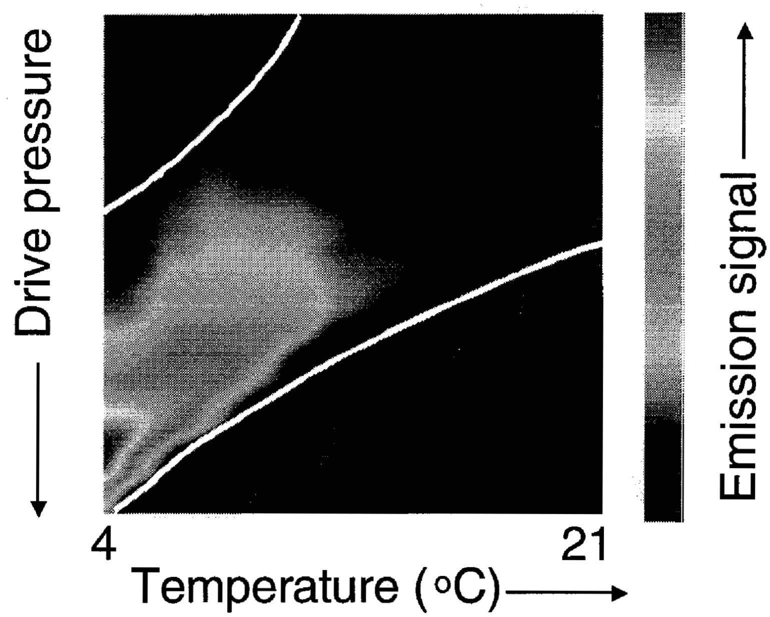

Figure 5. A map of the intensity of the sonoluminescence bubble as a function of acoustic drive pressure and water temperature. The limits of the SL process, indicated by the white lines, are seen to change with both temperature and density.

The mapping of the SBSL is important in that it allows us to study the trends and understand the interplay between the variables of temperature, drive pressure, and gas concentration. Indeed, in figure 6 we show the fact that the higher gas concentration give larger maximum bubble radius as one would expect. However, one can also see that the rations give substantially lower emission.

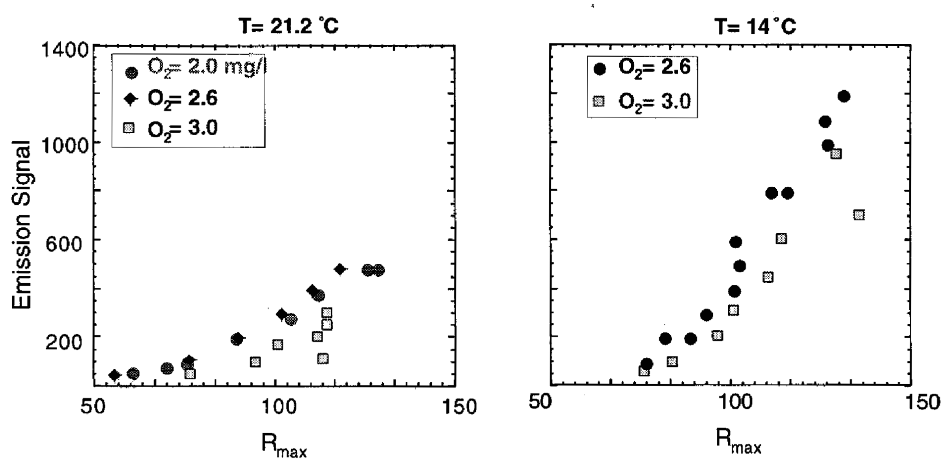

Figure 6. Two plots of the single bubble sonoluminescence signal on the abscissa versus the maximum bubble radius. On the left the cell was maintained at 21.2 C and three gas

concentrations plotted. Although the argon in the gas is the source of the emission the total oxygen represents the air concentration in the water. On the left the data is presented for a cell at 14 C showing that the bubble intensity goes up substantially for lower temperatures.

These results are more in depth data than has been seen. On the other hand, in figure 7 we show the new data that address the reason why lower temperatures yields bright SBSL signal. In figure 7 we see the SBSL emission intensity versus the maximum bubble radius for a single gas concentration and two temperatures. Note that at lower temperature the bubble is stable to higher drive pressure (correlated to the larger bubble maximum radius) , indicated by the data point shown in the region with the yellow background. However, this alone is not the answer as the bubble is already brighter for the same drive pressure (correlated to the data points where both temperature still provide SBSL emission).

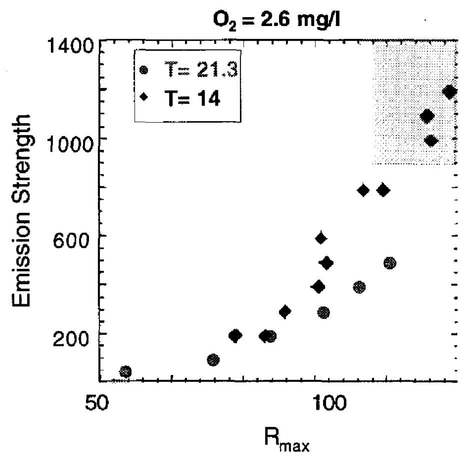

Figure 7. The variation of the SBSL emission at one gas concentration for tow temperatures versus maximum bubble radius.

Finally, we have performed the first experiments on the Schlieren measurements for the square sided cell and are now making the transition to the spherical cell. This effort will make it possible to map the dynamic behavior of the bubble radius as a function of time.

Potential Impacts

The importance of this work can be understood to as contribute to LLNL in two areas. First the area of sonoluminescence research has captured the attention of numerous students and remains in the public eye through media attention. We have, with the recent applications , managed to engage and utilize high quality students in the project. We have managed to connect through the students to a informal network of young people interested in sonoluminescence. These are young bright individuals that are being attracted to the laboratory and can participate in research in which they become fully involved. This is an exceedingly effective method for introducing researchers to LLNL and for gaining public awareness. This has been borne out in practice as we are continually attracting interested, excellent young researchers.

Second the sonoluminescence process accesses matter that is of itself interesting to LLNL. The experiments deal with the hydrodynamic collapse of a bubble in a liquid. The spherical nature of the collapse and the details of the simulations that need to be exercised are rigorous tests of the hydrodynamic codes. The fact that there is still no reasonable first principles solution of the source of the emissivity has led to studies that have impact on the equation of state of the gases in the bubble, as well as the study of water at extreme conditions. These studies, although potentially off the beaten ρ-T track, provide a means to improve our basic capabilities in a cost effective manner. Further, the experimental techniques that have been brought to bear on the problem,. Lasers timed with the RF driver now provide real-time images; The evolution of the experiments to perform laser based shadowgraphy, Schlerien, and interferometry will provide the hydrodynamicists with basic information on the ability of the codes to map out the implosion phase correctly.

Finally, the understanding of the light emission may provide a path for the use of this rather interesting phenomenon. This leads to the interesting possibility that the bubble may provide an insight in how to model the warm dense matter regime. Indeed this regime, roughly defined as the place in the ρ-T plane that is near solid density but too hot for normal condensed matter theory, while at the same time the density is too high for normal plasma statistical mechanical approaches. Here we have a laboratory experiment that generates conditions that may simply and reproducibly provide the opportunity to develop both data and modeling techniques.

Summary

Of course, there is much more interesting and important work in the area of SBSL research. In the area of experimental progress: First, we could complete the measurements using the Schlerein and interferometric techniques to develop a radius versus time set for various parts of the parameter space. Second, to further tap into the laboratory expertise in laser based diagnostic we could design a Thomson scattering setup to attempts measurements of the bubble temperature, electron density, and possibly the mean ionization state as a function of time. Third, we can develop a spectroscopic capability that can measure in the visible and in the near infra-red to address the predictions made by both the detailed spectroscopic models and by the simulations.

In the area of modeling improvements: First, one could model the high quality data from noble gas bubbles that has been presented by a group of collaborators at Yale. These data are of interest as all parameters have been carefully kept constant from on noble gas experiment to the next so it provide a rigorous test. Second, one should improve the spectral synthesis model to include the partition functions for the molecular species. This will allow studies of the bubble in the initiation phase of the problem and provide a test of the ionization balance in those case where we have warm dense matter.

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